



ISSN: 0067-2904

Improving Some Mechanical Properties and Thermal conductivity of PMMA Polymer by using Environmental Waste

Tagreed M. Al-Saadi^{*1}, Anaam W. Watan¹, Hanaa G. Attiya²

¹Department of Physics, College of Education for pure science Ibn -Al-Haithem, University of Baghdad, Baghdad, Iraq

²Department of Chemistry, College of Education for pure science Ibn -Al-Haithem, University of Baghdad, Baghdad, Iraq

Received: 20/7/2020

Accepted: 5/11/2020

Abstract

This study was achieved to satisfy two goals, the first of which is to treat an environmental problem represented by the disposal of date seeds, and the second is the use of these wastes to improve some mechanical and thermal properties of poly methyl methacrylate PMMA through strengthening different proportions of the powder of date seeds.

Particles of date seeds were used as a natural strengthening material for PMMA polymer, by mixing the matrix material (resin) with the hardener while still stirring continuously for a period of 10 min. After that, the samples of the reinforced material were prepared by adding the powder of date seeds, which is the reinforcing substance, with different percentages of weight fraction (0, 0.5, 1, 2, 3, 5 wt. %) and a grain size of <75 μm , while continuing to stir (10 min) for a second time. The composite samples were prepared by the Hand-Lay-up method and cut according to the standard ASTM. Thermal conductivity and some mechanical properties, such as impact strength, tensile strength, compressive strength, flexural strength, and hardness, were studied. An improvement was found in all properties at the reinforcement rate of 1-2 wt. %.

Keywords: Recycling waste, Dates seeds, PMMA resin, Thermal conductivity, Mechanical properties

تحسين بعض الخواص الميكانيكية والتوصيل الحراري لبوليمر PMMA باستخدام المخلفات البيئية

تغريد مسلم الساعدي^{*1}، أنعام وادي وطن¹، هناء غالي عطية²

¹قسم الفيزياء، كلية التربية للعلوم الصرفة - ابن الهيثم، جامعة بغداد، بغداد، العراق

²قسم الكيمياء، كلية التربية للعلوم الصرفة - ابن الهيثم جامعة بغداد، بغداد، العراق

الخلاصة

تهدف هذه الدراسة إلى تحقيق أمرين، أولهما معالجة مشكلة بيئية تتمثل في التخلص من بذور التمر، والثاني استخدام هذه المخلفات لتحسين بعض الخواص الميكانيكية والحرارية لراتنج بولي ميثا اكرليت PMMA من خلال تقويته بنسب مختلفة من مسحوق بذور التمر. تم استخدام مسحوق بذور التمر كمادة تقوية طبيعية لبوليمر PMMA عن طريق خلط مادة الأساس (الراتنج) مع مادة التقوية مع الاستمرار في التقليب المستمر لمدة (10 دقائق) حيث تم بعدها تحضير عينات المترابك البوليميرية وبكسور وزنية مختلفة من مادة التقوية

*Email: tagreedmm2000@gmail.com

(0، 0.5، 1، 2، 3، 5 %) وبجسم حبيبي بحدود $>75 \mu\text{m}$ مع الاستمرار في التحريك (10 دقائق) للمرة الثانية. تم تحضير العينات المترابطة بطريقة الصب اليدوي والتقطيع حسب المواصفة (ASTM). تمت دراسة الموصلية الحرارية وبعض الخواص الميكانيكية مثل مقاومة الصدمات وقوة الشد وقوة الانضغاط وقوة الانثناء والصلادة. وجد أن هناك تحسنا في جميع الخواص عند نسبة وزنية 1-2 % من مادة التقوية.

1- INTRODUCTION

An important environmental problem in many areas around the world is the accumulation of agricultural plant wastes, due to the lack of enough factories with complete efficiency of waste recycling. Date seeds are considered as one of the kinds of plant waste that is characterized by being dry, large-sized and light-weighted [1]. These wastes are a real problem when accumulating heavily near factories, especially when they are burned which causes a serious pollution of the environment, in addition to becoming a shelter for many harmful insects and rodents [2]. Therefore, it is necessary to consider how to use this waste, not only as animal feed, but also possibly as a reinforcement material after being mixed with a suitable polymer to become in the form of a composite that can be used for various industrial purposes.

Composites include two materials or more with different physical and mechanical properties to synthesize a new hybrid material that has characteristics not found in the original materials [3]. PMMA is a common polymer that is widely used as a polymeric material in different fields, including machinery, aerospace, architecture industries, and medical uses [4, 5]. In addition, PMMA is used in manufacturing removable denture base material [6]. Composites have special properties that enable them to be used in different structures and/or structural components without compromising their structural performance and durability. However, the use of natural materials in composites makes the structure more eco-friendly and economical [7, 8].

The objective of this research is to prepare PMMA /date seed composites with weight percentages of 0.5,1,2,3 and 5 wt. % of seeds as a powder in order to improve some mechanical and thermal properties, in addition to the efficient disposal of some environmental waste. On the other hand, such composite can be used in the dental industry, specifically in the bases for artificial dentures.

2- EXPERIMENTAL PART

2.1. Matrix material

In this research, PMMA (Evonik/ Germany) was used as a resin solution which is mixed with a hardener by a ratio of 4: 1 . This resin has a density of $1.17-1.20 \text{ g/cm}^3$. On the other hand, it is well known that the resin is transparent, thermoplastic, and strong.

2.2. Date seeds

Initially, the seeds were collected from one type of dates. Then, they were washed well by deionized water and dried in an oven at 50°C for 6 hr. After that, they were ground into a $<75 \mu\text{m}$ size powder.

2.3. Procedure of Composite Preparation

The mechanism of hand lay-up was utilized to prepare the sheets of PMMA / date seeds powder composites with dimensions of $20\text{cm}\times 20\text{cm}\times 0.4 \text{ cm}$. The date seeds were utilized in different weight fractions (0, 0.5, 1, 2, 3, 5 wt. %) with an appropriate quantity of PMMA. The samples were cut according to the required examination, as follows: impact strength as in ISO-179, tensile strength as in ASTM-D638, compressive strength as in ASTM-D695, flexural strength as in ASTM-D790, dielectric strength as in ASTM-D150, thermal conductivity as a disc (height 2mm, diameter 45mm), and hardness as a disc (height 5mm, diameter 20mm).

2.4. Measurements

1. Thermal conductivity

When there is a thermal difference between two surfaces, the heat will pass from the high-temperature surface to that of low temperature, a phenomenon that is known as thermal conductivity. On this basis, thermal conductivity can be defined as the rate of heat flow across the unit area during the time unit when there is a thermal gradient between two surfaces of one degree Celsius. Thermal conductivity is different depending on the material (solid, liquid, gaseous), according to which the material is classified as a dielectric or heat conductor [9].

Thermal conductivity in metallic materials depends on the transfer of free electrons, whereas in composite materials it is adopted where the composite materials show a strong conductivity, as in the

fiber orientation (through the thickness) or a weaker conductivity in the longitudinal direction of the fiber. In general, thermal conductivity of resins is increased after fiber reinforcement and this increase is expected due to the fiber capacity as a whole for thermal conductivity compared to the base material [10].

The coefficient of thermal conductivity was measured by utilizing lee's disk (Griffin and George / England), based on the following equations [11]:

$$K \frac{(T_2 - T_1)}{d_s} = e \left[T_1 + \frac{2}{r} \left(d_1 + \frac{d_s}{2} \right) T_1 + \frac{d_s - T_2}{r} \right] \dots \dots \dots (1)$$

In addition, the amount of thermal energy (e) was determined by [12].

$$I V = \pi r^2 e (T_1 + T_3) + 2\pi r e \left[d_1 T_1 d_s \frac{(T_1 + T_2)}{2} + d_2 T_2 + d_3 T_3 \right] \dots (2)$$

K: the coefficient of thermal conductivity (W / m . °C).

e: the amount of thermal energy that passes through a disk material's unit area per second.

d₁, d₂: disk's thickness (mm). d_s: sample's thickness (mm).

2. The Mechanical Properties of PMMA/Date Seeds composite

The general and engineering uses of composite materials depend largely on their mechanical and physical properties, such as tensile strength, elasticity, and elongation, as well as resistance to heat and environmental conditions, such as humidity and sunlight. All these properties depend heavily on the molecular structure of the resin along with its molecular weight and molecular forces. These properties also depend on the reinforcing materials and the additives such as fillings and plasticizers [13]. The following characteristics are discussed in this research.

- Impact Strength

Impact strength expresses the material's ability to resist breakage under a sudden load and is a measure of the strength of the material. Impact strength can be expressed by the following formula [14]:

$$I. S. = \frac{U_f}{A} \left(\frac{KJ}{m^2} \right) \dots \dots \dots (3)$$

where A is the of cross-sectional sample's area in m² and U_f is the energy of fracture (KJ).

- Tensile strength

Tensile strength is a measure of the material's ability to resist static forces that attempt to pull and break the material. The tensile strength can be expressed by the following formula [4]:

$$T. S. = \frac{F}{A} (MPa) \dots \dots \dots (4)$$

where A is the cross-sectional area of the sample in mm² and F is the applied force (N).

- Compressive Strength

This resistance measure shows the extent to which the material is tolerated when it undergoes a static compression load before it is broken, usually measured by the Mpa unit. The high values of tensile strength indicate the greater cohesion forces between the molecules of the material. The compressive strength can be expressed by the following formula [4]:

$$C. S. = \frac{F}{A} \left(\frac{N}{m^2} \right) \dots \dots \dots (5)$$

where A is the cross-sectional area of the sample (mm²) and F is the applied force (N).

- Flexural Strength

This property is a measure of resistance to bending and can be defined as the maximum static load that can be affected on the test sample before being broken, measured in the unit of MPa. The flexural Strength can be expressed by the following formula:

$$F. S. = \frac{3Pl}{2bd^2} (MPa) \dots \dots \dots (6)$$

where l is the distance between the two support points, b and d are the sample dimensions, and P is the applied load [14].

- Hardness

Hardness is defined as the material resistance to scratching or penetration. There are several different global standards for determining the hardness of plastic materials, the most common of which is the hardness of the Shore D. The penetration occurs at a slow rate on the surface of the sample during the effects of force, leading to a local creep. After the vanishing of the influencing force, a relatively slow

recovery occurs, which changes the dimensions [15]. The hardness of all samples was examined according to the standard specification (TH 210) of Shore (D) device.

- Dielectric Strength

The dielectric strength can be measured by the electric field (E_{br}), such as the field at which the dielectric material fails and collapses, according to the following formula [4]:

$$E_{br} = V_{br} / h \dots\dots\dots (7)$$

where V_{br} is the maximum voltage applied to the dielectric and h is the thickness of the dielectric material. Dielectric strength or breakdown units of measurement are kV / cm or kV / mm.

The reasoning for the occurrence of a breakdown in the insulation is the occurrence of a hole, smelting, or burning in the dielectric material.

3- RESULTS AND DISCUSSION

1. Thermal Conductivity

Figure-1 shows the relationship between the thermal conductivity coefficient and the reinforcement materials (date seeds) for different fractions.

Increasing the fractions of date seeds in the PMMA matrix increased the thermal conductivity of the composite material up to 1 wt. % due to the increase in the number of fine spaces in the compound. This is a result of the greater amount of bonding area between the date seeds and the polymer matrix, when the thermal energy transfers through the matrix material by the vibration of the material. After this ratio, the high percentage of date seeds powder may lead to a lack of cohesion and the presence of pores and air voids that lead to a reduction of the thermal conductivity coefficient, as it is known that the air has a very low thermal conductivity coefficient of 0.026 (W / m K) at 25°C . On the other hand, this leads to a difference in the area of the interface in which the energy transfers, which will block the transmission of heat energy and thus reduce the value of the transferred heat energy, leading to lower thermal conductivity values [16, 17]. On the other hand, the addition of date seeds to the polymer works to increase the thermal conductivity significantly. The reason for this is that the fine date seeds fill the gaps between the layers of the polymer, in addition to the fact that date seeds provide a good adhesion to the polymer [11].

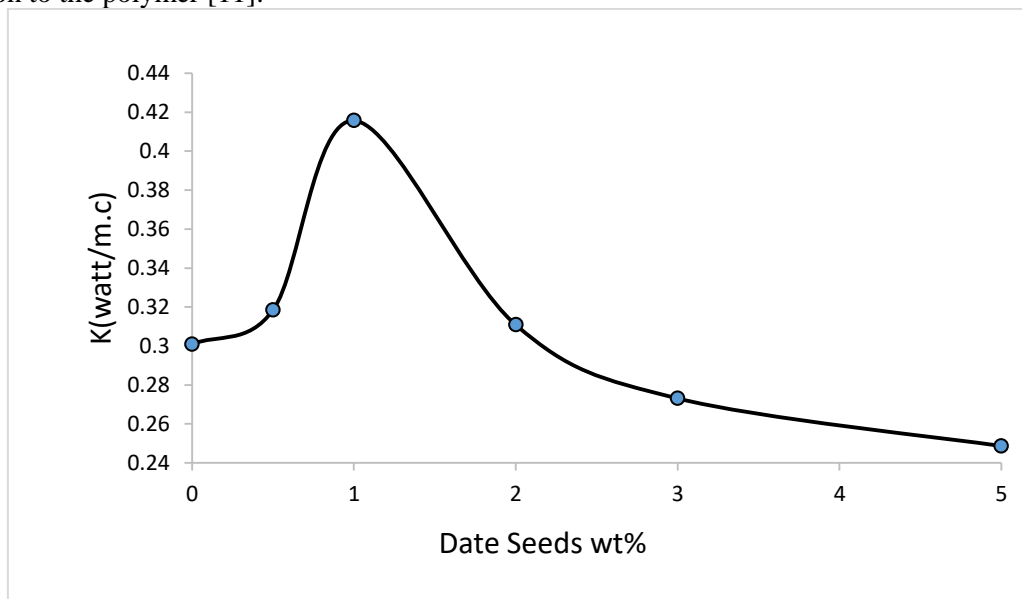


Figure 1-The relationship between thermal conductivity and wt. % of date seeds

2. Impact Strength

Figure-2 explains the variation of impact strength with the percentage of filler content of date seeds in reinforced PMMA composites. For the sample with 1 wt. % seed content, we can notice the maximum value of 5.83 KJ/mm², with an increase of about 59.7%. After the reinforcement process, the value of impact strength decreased. Accordingly, when the ratio of the date seeds increases, the poor interfacial adhesion appeared among the reinforcement material and the PMMA matrix.

The impact strength of composites is controlled fundamentally by the capability of the natural fillers to absorb energy, which can stop crack propagation. The other factor is the good interfacial bonding

which produces micro-spaces between the natural fillers and the matrix, resulting in difficult crack propagation [6]. When one or both of these factors are absent, this leads to the appearance of cracks in micro dimensions at the points of impact, which reduces the impact strength. This happens because of the insufficiency of the reinforcements to prevent the propagation of cracks, resulting in the decrease of the impact strength [18, 19].

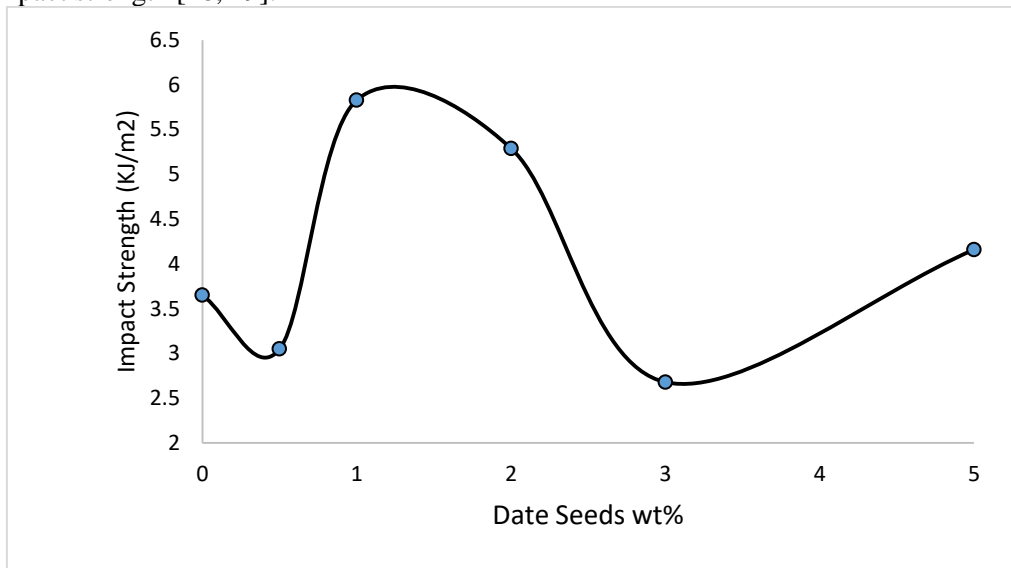


Figure 2-The variation of impact strength with wt. % of date seeds

3. Tensile Strength

The response of the materials to tensile strength is shown in Figure-3. Treatment of the polymer with fillers makes it more cohesive, resulting in improved bonding between the various constituents. Both date seeds and PMMA composite gave better tensile strength compared to the pure PMMA. The use of date seeds in different percentages as a filler gave more enhancement in tensile strength than pure PMMA samples. The highest tensile strength of 10.93 MPa was obtained for composites with 1wt. % filler of date seeds, which is an enhancement of about 88.7 % over composites of lowest filler. Further addition of date seeds and PMMA to the composites resulted in the reduction of tensile strength, which is also observed in the flexure behavior. Above 1wt % for filler loading, the tensile strength decreased, possibly due to a lack of compaction between the reinforcements and the polymer matrix and inhomogeneity in distribution [8].

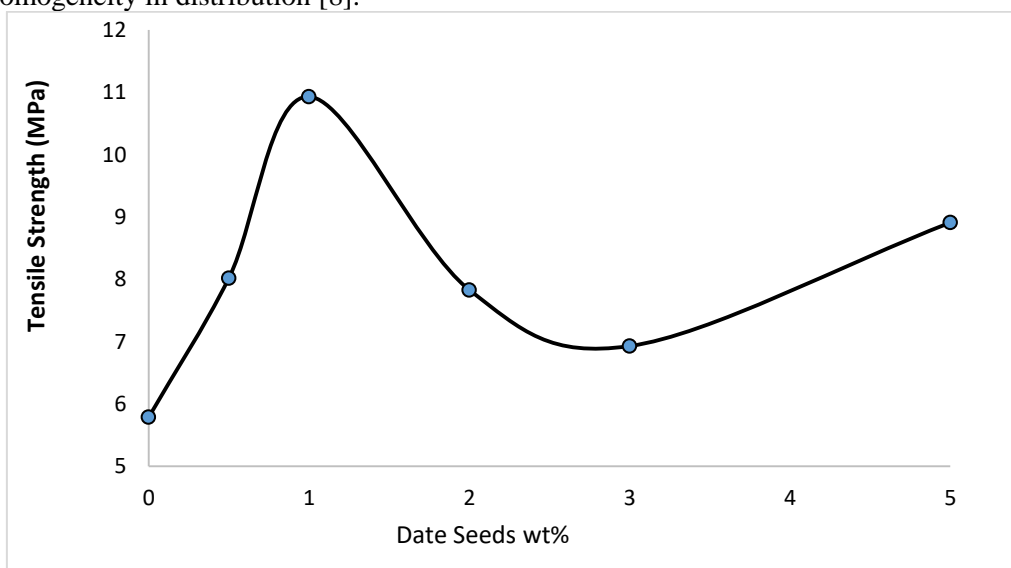


Figure 3-The variation of tensile strength with wt. % of date seeds

4. Compressive Strength

Figure-4 exhibits the influence of the date seeds fraction and PMMA at different percent loading on the compressive stress of the PMMA matrix. Figure-4 displays the increase in compressive strength when date seeds powder was added to PMMA matrix, which may be due to the fact that PMMA has high compressive strength, along with the good interface between the filler and matrix. The compression strength for the composite gradually decreased when the filler content was beyond 1% wt. (12.6% MPa). This indicates that the lower degree of particles-polymer interaction occurs at higher filler contents, because of the agglomeration of the powder of date seeds when the content of filler increased [20].

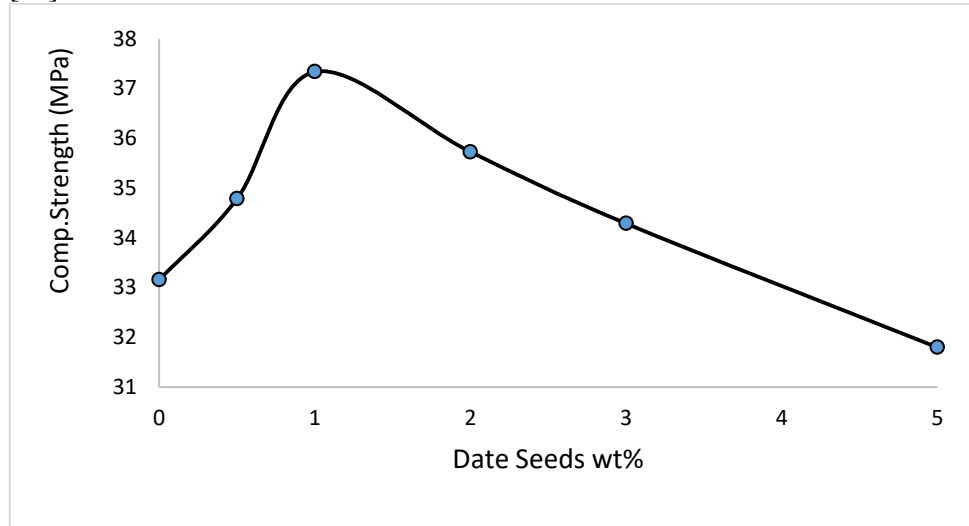


Figure 4-The relationship between Compressive Strength and wt. % of date seeds

5. Flexural Strength

The flexural strength of polyester composites relies certainly on the composite's microstructure and the bonding of interfacial between the matrix and the reinforcement material [21]. Flexural strength of the composites is presented in Figure-5. It can be observed that the addition of date seeds increased the flexural strength of the composites by about 101.7% for 1wt. % sample. When the pure polymer was compared with the reinforced composite, it can be observed that there is a change in the value of flexural strength from 46.91 N/mm² to 94.63 N/mm², which is the highest value among all composites at 1wt %. This is because of the strong interfacial adhesion/bonding among the particles of the date seeds and the matrix, which enhances the transfer of load [18]. The subsequent reduction in flexural strength is because of the agglomerate formation at advanced ratios of the reinforcement material. The lowest value of flexural strength was 94.63 MPa, which was obtained from the composite with 1% date seeds filler, as shown in Figure-5.

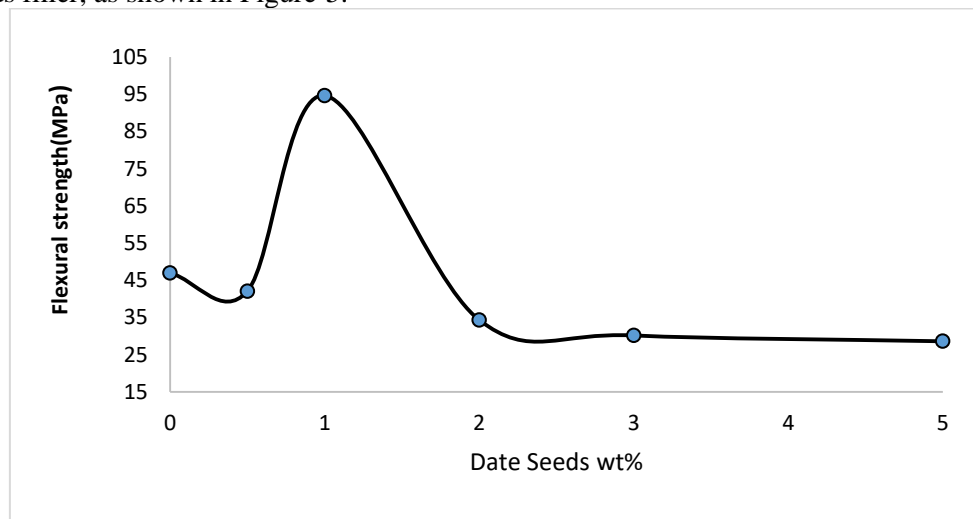


Figure 5-The variation of flexure strength with wt. % date seeds

6. Hardness

Figure-6 illustrates the variation of hardness (Shore D) with different weight percentages of date seeds added to the composites. The hardness of the composite increased with the increase in filler content of date seeds. From this figure, it can be shown that the hardness increased with increasing the addition at 3 wt. % (4.2% Shore D), which displayed the highest hardness value compared to all other investigated samples, as shown in Figure-6. This observation may be due to better compaction between the reinforcing materials and possibly due to the inhomogeneity of the samples resulting from poor manual compaction of the sample. Generally, the addition of fillers increases the hardness of composite materials, especially if well compacted.

However, the presence of date seeds was observed to significantly affect the hardness value. This observation is in agreement with the results of other studies [22, 23]. In addition, the PMMA /date seeds composite showed higher hardness compared to pure PMMA, because of the strong bonding between PMMA and date seeds and the amorphous nature of PMMA. It can be deduced that the hardness of a composite depends on the distribution of the reinforcing material in the matrix. Shore hardness is a measure of the resistance of a material to deformation [10]; a higher number indicates greater resistance.

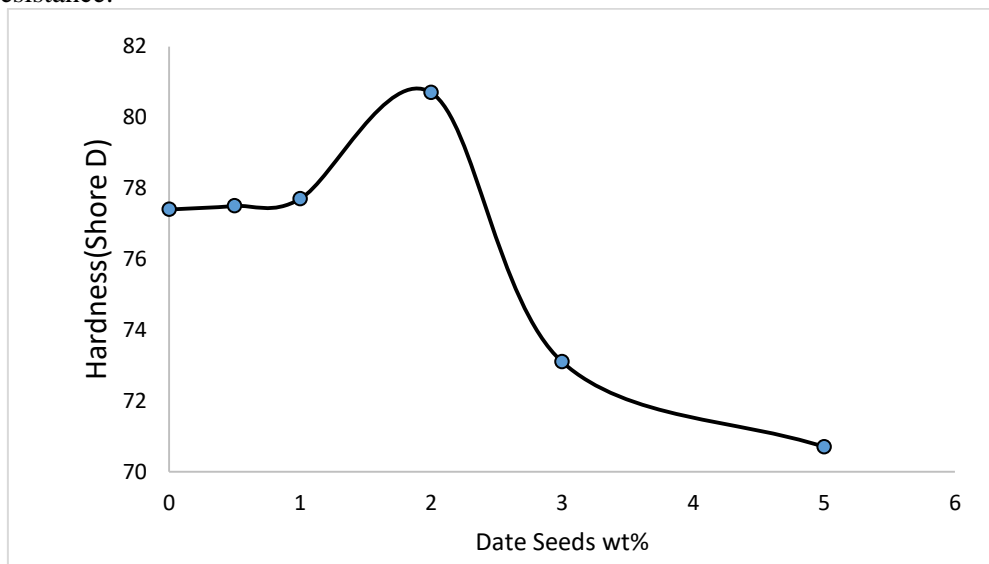


Figure 6-The variation of hardness (Shore D) with wt. % date seeds

7. Dielectric Strength

Figure-7 reveals the response of the materials to breakdown voltage after treatment of the polymer with filler, which provides more cohesion that result in improved bonding between the various constituents. Both date seeds and PMMA composite gave better breakdown voltage compared to the pure PMMA, as shown in Figure-3. The use of date seeds as a filler in different percentages gave more enhancement in breakdown voltage than pure PMMA samples. The highest breakdown voltage of 25.76 kV/mm was obtained for composites with 3wt. % filler of date seeds, which is an enhancement of about 18.5 % over composites of lowest filler. Further addition of date seeds and PMMA to the composites resulted in the reduction of breakdown voltage, which is also observed in the flexure behavior. Above 1wt % of filler loading, the breakdown voltage decreased, possibly due to a lack of compaction between the date seeds and the polymer matrix [8]. In most of the polymers of PMMA composite, the bundle structure was formed because of the agglomeration of filler particles at the nearness of convenience [24]. From this, it is evident that high percentages of the date seeds do not achieve good results in dielectric strength [11].

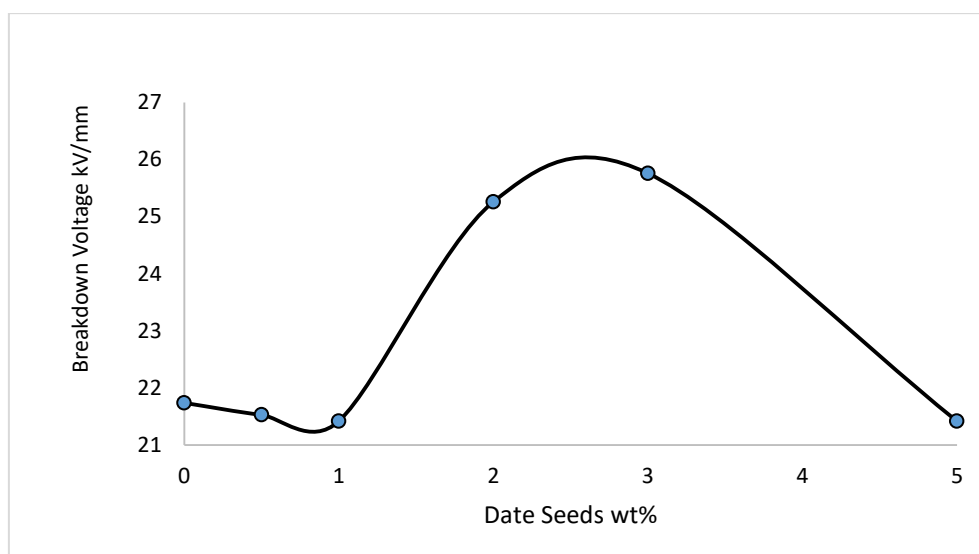


Figure 7- The relationship between breakdown voltage and wt. % date seeds

4. Conclusions

This study could treat the negative effects of wastes by integrating them into applications that serve environmental sustainability. Economically, it is possible to use powders of date seeds instead of the synthesis compounds in many applications, including dental industry, specifically in the bases for artificial dentures, due to good mechanical properties and the low cost of manufacturing. Moreover, the use of these seeds improved some of the mechanical and thermal properties of the polymeric composite.

Date seeds caused an enhancement in the mechanical and thermal properties in addition to the dielectric strength in PMMA / date seeds composites. The increases to the highest values happened at a weight fraction of 1-2 wt. % due to the strength of the bond between the matrix material and the date seeds and the uniform distribution of the reinforced material particles at low percentages of filling. On the other hand, the decreases in mechanical properties and thermal conductivity at a high percentage of date seeds were due to the heterogeneous distribution for particles inside the PMMA.

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